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Comparative Study of CO_2 Capture in the French Context

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Abstract

This paper presents a summary of technical-economic studies allowing to evaluate the production cost of electricity derived from coal and gas power plants with the capture of CO_2, and the cost per ton of CO_2 avoided, in the French context. Three systems were studied: an Integrated Gasification Combined Cycle (IGCC), a conventional combustion of Pulverized Coal (PC) and a Natural Gas Combined Cycle (NGCC). For the IGCC, two gasification types have been studied: a dry coal gasification at 27 bars, and a technology based on gasification of a coal - water mixture (slurry) that can be compressed to 64 bars (GE/Texaco slurry type).

Keywords: CO_2 capture; pre-combustion; post-combustion; oxy-combustion; coal; power plant; avoided CO_2

Nomenclature

IGCC:	Integrated gasification combined cycle
IGCC-1200:	IGCC of quench type delivering 1200 MWe as gross power output
IGCC-300:	IGCC of radiant type delivering 300 MWe as gross power output
NGCC:	Natural Gas Combined Cycle
NGCC-480:	advanced NGCC delivering 480 MWe as gross power output
PC:	Pulverised coal power plant
PC-1200:	PC delivering 1200 MWe as gross power output
HHV:	High Heating Value (total calorific value of fuel)
LHV:	Low Heating Value (HHV less latent heat of the water of combustion)
ASU:	Air separation unit
MEA:	methylethanolamine
DEA:	diethanolamine
MDEA:	methyldiethanolamine
Selexol™:	process using dimethylether of polyethylene glycol (DMPEG)
Syngas:	synthetic gas produced by the gasification

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1. Introduction

The industrial feasibility of separating CO_2 contained in a gas mixture no longer needs to be demonstrated: for example, the ammonia industry uses many hydrogen production units derived from natural gas, in which the final step is CO_2/H_2 separation. Similarly, in the field of natural gas production, excess CO_2 contained in the gas in some deposits has to be extracted so that methane can be marketed. In the case of exhaust gases from electrical power stations, other handicaps include firstly the presence of impurities, and secondly the dilution of CO_2 by nitrogen in the combustion air: in general, CO_2 only represents 5% (for the NGCC system) to 15% (for the PC system) of the volume of exhaust gases. If it is required to separate the CO_2 efficiently, it is obviously tempting to remove the nitrogen from the air in order to make the capture process easier. For the IGCC process, this option was necessary even before the emergence of the CO_2 / greenhouse effect problem, mainly for economic reasons related to the reduction in the volume of installations and therefore the resulting savings in materials, the savings being particularly important when the gasification pressure is increased. For other processes, the use of oxygen as an oxidant requires that exhaust gases should be recycled to maintain conventional temperature levels and thus avoid the need to completely change the design of all combustion systems (boilers for PC and combustion chambers for NGCCs). However, note that for NGCC the problem of the expansion turbine arises because it is not adapted to a working fluid composed mainly of CO_2.

2. Different CO_2 capture processes

Our studies [1- 4], present more technical details as well as bulky bibliography on CO_2 capture. The present paper considers four different fossil fuel based power generation systems: the radiant type IGCC (IGCC–300), the slurry type IGCC (IGCC–1200), the supercritical PC-1200 and the NGCC-480. It can be considered that these are the most representative systems of the installations that will be used to replace existing fleets: the radiant type IGCC remains the reference (technology similar to that proposed by Shell), but the Slurry type process is included because its production cost is lower than the radiant type, despite its lower efficiency. The PC-1200 can be constructed at the present time and there is a significant benefit from the scale effect and its efficiency is better than the subcritical that is not presented herein. Three main methods can be envisaged for the capture of CO_2 with these three systems:

a) **pre-combustion capture**: to capture CO_2 in a synthesis gas after conversion of CO into CO_2;
b) **post-combustion capture**: to capture CO_2 in the exhaust gases once the fuel has been fully burned with air;
c) **capture in oxy-combustion**: consisting of combustion in oxygen with recycling of exhaust gases (therefore composed mainly of CO_2 and water) and purification of the CO_2 flow, to eliminate incondensable gases.

Various processes can be envisaged for separation of the CO_2 contained in a gas mixture. They are based on chemical, physical or hybrid absorption, adsorption, membranes separation or cryogenic separation [5-10]. However, only physical and chemical (or hybrid) absorptions and oxy-combustion associated with purification of CO_2 (usually cryogenically) appear to be suitable for use in high capacity power stations, but the choice of the "best" absorbent remains a very open question and oxy-combustion still has to be demonstrated on the industrial scale and several years of research and development will probably still be necessary (probably 15 to 20 years). Chemical processes based on primary amines such as MEA (monoethanolamine) are preferred when the partial pressure of CO_2 is genuinely very low (less than 1 bar), while chemical processes based on tertiary amines such as MDEA (methyldiethanolamine) are preferred when the partial pressure of CO_2 is slightly higher; MDEA is actually easier to regenerate under these conditions and its lack of reactivity is compensated by the addition of a kinetic absorption activator such as piperazine. The absorption capacity of amines increases with the partial pressure of CO_2 and begins to saturate at a partial pressure of 8 bars; beyond this limit, the performance of physical absorption improves and increases linearly with the partial pressure of CO_2 (Henry's law) [11].

2.1. Application to the capture of CO_2 for different thermal power station systems

Not all systems are compatible with all capture methods: the IGCC system can only be envisaged with the pre-combustion capture method [7] (the high pressure is used to separate CO_2 by physical absorption), the PC system

can be adapted to post-combustion capture and oxy-combustion, and the NGCC system can be adapted to the three capture methods, even though the pre-combustion capture option (reforming of methane and then capture of CO_2 on synthetic gas after conversion of CO into CO_2) is economically fairly expensive compared with the other two methods (this method should only be attractive in special cases, for example when hydrogen is necessary for a refinery).

Sensitivity and optimization studies were carried out for each of the options envisaged for the three systems. For post-combustion and pre-combustion capture, we considered the optimization of the CO_2 content in the solvent, the theoretical number of plates in absorption and regeneration columns, the solvent flow, the reflux ratio in the regeneration column, the regeneration heat and obviously the CO_2 capture ratio itself. For oxy-combustion, we considered optimization of the level of purity provided by the Air Separation Unit (ASU), the degree of purity of the CO_2 produced, the degree of leak tightness of the exhaust gas circuit (in the case of PC) and a study on the influence of these parameters on the CO_2 capture rate.

2.2. Basic assumptions

The results presented herein are all based on the same ISO ambient conditions (15°C, 1.013 bars and 60% relative humidity). The pressure and the relative humidity have a significant impact on the performances of combustion turbines, and the ambient temperature imposes a vacuum at the condenser and therefore controls the performances of steam turbines. Concerning fuels, the coal used is a standard international quality steam coal (LHV ~ 26 MJ/kg, 7% humidity, 15% ash and 1% sulfur). The annual operating times used (operation in base) are 8000 hours for NGCC, 7800 hours for PC and 7500 hours for IGCC. Installations are amortized over 30 years for all systems. The maintenance item for NGCC was reinforced to justify this minimum operating duration. For availability, we assumed 92% for NGCC, 90% for PC and 85% for IGCC.

The economic estimates in our studies have been produced on consistent bases using exactly the same assumptions for each of the different systems and the different capture processes; they can thus be used to make fairly accurate comparisons between the different systems studied (except for uncertainties in the estimates that are far from being negligible); on the other hand, their absolute values may be quite inaccurate compared with 2008 prices (for example, due to the fast increase in the price of metals or the price of fuel); It's why the absolute costs are not given in this paper, but only relative costs to Pulverized coal power plant without CO_2 capture which (used as reference). All relative costs mentioned in this work exclude transport and storage costs of CO_2. For engineering and management of installations, we increased equipment costs by 8% for PC and NGCC, and by 10% for IGCC. We also allowed for contingencies equal to 5% for PC and NGCC, and 8% for IGCC. The results presented herein are based on a discount rate of 11%, considered to be the most probable in the medium and long term and construction times were fixed at 2 years for NGCC, 4 years for PC and 5 years for IGCC. We also assumed parity between the American Dollar and the Euro.

We used the following reference values for the purchase prices of coal and natural gas:
- for coal: 60 Euros/tonne, equivalent to 2.26 Euros/GJ HHV;
- for natural gas: a price of 6.29 Euros/GJ HHV.

3. Comparison between different capture processes

The calculations (see Table below) show that the highest efficiency with capture is obtained for NGCC with post-combustion capture thanks to the relatively high efficiency of NGCC without capture (50% with capture and 60% without capture). The next highest is the oxy-combustion in PC, with an efficiency of almost 35% compared with 45% without capture, and then IGCC-Puertollanowith pre-combustion capture with an efficiency of the order of 33.5% compared with 44% without capture. The lowest efficiency is obtained with post-combustion capture in PC, equal to 30% which is 15 points less than PC without capture (if MEA is used). Therefore, from a purely energetic point of view without considering economic figures, it would be recommended to only consider pre-combustion capture in IGCC, post-combustion in NGCC and oxy-combustion in PC. Nevertheless, post-combustion capture in

PC, although its efficiency seems to be lower than oxy-combustion PC, is now seen inevitable if it is required to be able to retrofit coal-fired power stations that will be built between now and 2020 (which accounts for more than 1000 GW). For NGCC, oxy-combustion is not a possibility because not all current combustion turbines are suitable for this CO_2 capturing process; oxy-combustion of natural gas could only become technically viable if innovative cycles are adopted, and provided that they can be brought into an industrialization phase in order to reduce the cost.

	Reference Power Plants				Pre-combustion			Oxy-combustion		Post-combustion	
	IGCC-300	IGCC-1200	NGCC-480	PC-1200	IGCC-300	IGCC-1200	NGCC-480	NGCC-480	PC-1200	NGCC-480	PC-1200
	w/o capture	w/o capture	w/o capture	w/o capture	Pre-comb.	Pre-comb.	Pre-comb.	Oxy-comb.	Oxy-comb.	Post-comb.	Post-comb.
	Mass balance										
Fuel flow rate (Tonne/heure)	98	349	59	346	112	378	76	59	340	59	346
Thermal Power (MWth)	742	2 500	800	2 473	852	2 704	1 038	800	2 430	800	2 473
Net Efficiency (LHV)	43,9%	41,4%	59,7%	44,6%	33,5%	32,5%	45,8%	47,8%	34,7%	49,7%	30,0%
Net Power output (MWe)	**326**	**1 034**	**478**	**1 103**	**285**	**878**	**475**	**383**	**843**	**398**	**742**
Hours/year	**7 500**	**7 500**	**8 000**	**7 800**	**7 500**	**7 500**	**8 000**	**8 000**	**7 800**	**8 000**	**7 800**
CO2 emitted (tonne/heure)	240	822	158	817	25	141	23	16	80	17	86
CO2 emitted (kg/kWh)	0,735	0,795	0,332	0,741	0,088	0,160	0,049	0,042	0,095	0,044	0,115
CO2 captured (tonne/heure)					237	797	182	142	722	141	717
CO2 captured (kg/kWh)					0,831	0,908	0,383	0,371	0,856	0,354	0,966
captaure rate					85,51%	85,01%	88,61%	89,82%	89,99%	88,98%	89,33%
CO2 avoided (kg/kWh)					**0,594**	**0,634**	**0,282**	**0,290**	**0,646**	**0,288**	**0,625**
	Cost										
Capital Cost (relatively to PC)	**122**	**101**	**40**	**100**	**185**	**164**	**130**	**104**	**169**	**83**	**178**
Investment	**55**	**46**	**17**	**43**	**84**	**74**	**55**	**44**	**73**	**35**	**77**
Fuel	**43**	**46**	**95**	**43**	**57**	**58**	**124**	**119**	**55**	**114**	**63**
O&M	**19**	**19**	**11**	**14**	**22**	**22**	**13**	**11**	**20**	**13**	**25**
Production Cost (relatively to PC)	**117**	**111**	**123**	**100**	**163**	**154**	**192**	**174**	**148**	**162**	**165**
Cost of avoided CO2 (relatively to post-combustion in PC)					**73**	**65**	**234**	**168**	**71**	**130**	**100**

The lowest production costs (see Table above) are obtained with oxy-combustion PC (148% of PC without capture) and IGCC-slurry with pre-combustion capture). The production cost of radiant type IGCC with capture in pre-combustion is of the same order of magnitude as NGCC with post-combustion capture (162%).

On the other hand, the lowest costs per tonne of CO_2 avoided are obtained with pre-combustion capture in IGCC-slurry (65% of the cost of CO_2 avoided in PC with post-combustion capture), oxy-combustion in PC (71%) and the radiant type IGCC with capture in pre-combustion (73%) (see Table above). The highest cost per tonne of CO_2 (234%) is obtained using pre-combustion capture in NGCC.

4. Limitations of the different capture processes

In this chapter, without attempting to be exhaustive, we will summarize the assumptions that form limitations to the conclusions of our evaluations, limitations concerning technical and economic aspects

4.1. Technical limitations

Problems remain with oxy-coal combustion boilers, hydrogen turbine for IGCC with CO_2 capture, degradation of amines, separation of incondensable gases in oxy-combustion (what about impurities in a cryogenic system?), etc. These main limitations are described below.

4.1.1. Limitations on the industrial feasibility of processes

Although there are some industrial references for most capture processes for coal power stations that have been mentioned, they are not for identical applications: for example, chemical absorption has been done on an industrial scale for a long time (ammonia and fertilizer industries, separation of CO_2 from natural gas). Chemical absorption will have to satisfy two criteria before it can be used to capture CO_2 in exhaust gases from coal fired power stations,

related firstly to its high energy demand for regeneration of the absorbent, and secondly degradation of the absorbent by impurities contained in the exhaust gases to be treated.

The impurities will also create a problem for the cryogenic purification system associated with oxy-combustion: corrosion, formation of crystals, etc. More generally, oxy-combustion has been used by glassmakers for a long time but on very different furnace technologies, with much higher temperature levels and in the absence of exhaust gas purification systems, and it will create operation and safety problems for the electricity producer; it will be necessary firstly to adapt to a new operations culture that imposes strict control over safety with the use of pure oxygen as an oxidant, and secondly to solve combustion, corrosion, leak tightness, CO_2 purification problems, and no doubt other problems not even known at the present time.

In addition to safety problems related to the use of pure oxygen and control over highly toxic gases (CO and H_2S), IGCC with capture requires good control over combustion of hydrogen in combustion turbines. Although most manufacturers do make hydrogen combustion turbines at the present time, they remain modest in size and long term experience with them is quite limited. Obviously, the main problems that arise with hydrogen combustion, apart from safety problems, are flame stability (flame extinguishing and flashback), and NO_x emissions that may quickly become excessive.

Thus and in general, CO_2 separation techniques will introduce chemistry into electrical power stations to a varying degree, which in particular will impact its classic operation (change of "culture") and the risk management, as a result of the large quantities of chemicals involved in these processes.

4.1.2. Limitations on the industrial feasibility of "Capture ready"

All systems that envisage CO_2 capture require a significantly larger footprint than standard reference power stations. In addition to the space to be set aside for the CO_2 compression unit, the footprint occupied by the capture unit will obviously depend on the process used for CO_2 capture and the size will vary depending on the reactivity of the solvent used. For example, processes based on primary amines such as MEA will require a smaller footprint due to the high reactivity of primary amines and therefore will be able to operate with smaller absorption columns. Conversely, processes based on tertiary amines or other less reactive amines will require a larger footprint. This area could be optimized by integrating the different elements of the post-combustion capture process into the PC power station separately. Obviously, this should be examined case by case but in any case it will be done at the price of increased complexity in operation and also a higher pipework cost. The different capture processes are not "equal" faced with the possible necessity of making "capture ready". Apart from the problem of available space to accommodate the capture unit, the addition of some technical provisions will have to be anticipated.

For example for IGCC, the CO conversion unit will have to be included from the beginning, even if the power station does not include CO_2 capture: this is necessary for better circulation of synthetic gas in the pipes and equipment. This equipment cannot be adapted so that it can be used for a synthetic gas (CO, H_2) typical of IGCC without capture, or for a synthetic gas (CO_2, H_2) or for almost pure hydrogen specific to IGCC with capture. Therefore, there will be an extra investment and production cost for IGCC during the operation phase without capture of CO_2.

Similarly, for post-combustion capture for which large amounts of steam are drawn off, it will be necessary to design a flexible steam turbine system that can operate with maximum flows and with very low flows of low pressure steam.

Another problem that arises for oxy-combustion is multi-fluid burners: coal + air and coal + oxygen + CO_2.

4.1.3. Limitations on the quality of CO_2 for transport and storage

The presence of large quantities of incondensable gases in the CO_2 flow transported by pipeline in the supercritical state can cause vibrations and shock loads, which can cause mechanical damage. However, no known

study has attempted to specify critical concentrations of these incondensable gases, although it would appear that there is general agreement that the minimum required purity of CO_2 is 96%. All that is agreed upon unanimously is the concentration of water and most works published on this subject agree upon a fixed limit of 20 ppm by mass (50 ppm by volume). In fact, it is technically and economically difficult to dry the gas below this limit and it is believed that this limit will avoid corrosion problems. This assumes perfect circulation of the CO_2 flow in the pipe and therefore that there will be no accumulation of water in the pipe. Corrosion problems will certainly occur if this is not the case (for example the presence of interstices inside the pipe). Dangers related to a pipe break and to a massive leak of CO_2 may be serious because cold CO_2 is heavier than air, causing the risk of anoxia. Note that in this respect, an odorant can be added to CO_2 to make it easier to detect in the case of a leak, like what is done for natural gas. Other known limitations relate to SO_x, that must be limited to less than 10 ppm if CO_2 is to be used for enhanced oil recovery (EOR).

4.1.4. Limitations on resources

Capture of CO_2 consumes large amounts of energy, which can cause problems in a context admittedly marked by the greenhouse effect problem, but also by the beginning of rarefaction of fossil primary sources. Paradoxically, to avoid the greenhouse effect, we are obliged to consume more fossil fuels because efficiencies are reduced to be no better than they were at the beginning at the 20th century!

In addition to the primary energy consumed in the capture processes (amines regeneration, technical oxygen production, CO2 purification…), CCS consumes large quantities of water in a context in which water resources are becoming valuable. Admittedly, dry cooling technologies lose very little water, but they significantly reduce the efficiency of the power station. Therefore, residual heat from the power station is evacuated with the wet technology in closed circuits bringing ambient air and cooling water into contact, to avoid even higher losses of efficiency. With this technology, 2 to 2.4 liters of water per steam kWh are lost by evaporation. This is why the PC process consumes the largest quantity of water, because all the electricity is generated with steam that has to be condensed to be recycled. IGCC and NGCC consume lower quantities of water than PC because almost two thirds of the kWh are produced by the combustion turbine for which heat is evacuated with combustion exhaust gases.

4.1.5. Limitations on management of waste

Alcanolamine solutions are some of the most frequently used chemical solvents in acid gas absorption processes. Monoethanolamine (MEA) and diglycolamine (DGA), primary amines, diethanolamine (DEA), secondary amine, and methyldiethanolamine (MDEA), tertiary amine are the most frequently used amines. MEA is the most reactive amine; it reacts quickly with acid gases, and enables high quality separations in terms of the purity of separated products. However, impurities present in the gas to be treated such as SO_2 can cause the irreversible formation of stable degradation products. Similarly, an irreversible formation of solid carbamate could occur if the temperature exceeds 120°C during regeneration of the solvent, due to bad control. These degradations reduce the capacity of the solvent to absorb CO_2, increase its viscosity and cause a tendency to foaming. Furthermore, degradation products are highly toxic and a first bibliographic analysis shows that the only means of eliminating them are incineration or burial. Therefore, the treatment of waste / by-products will be significantly worse than they are for reference power stations without capture. The number of absorption / desorption cycles during a year of operation will be very high: several thousands or tens of thousands depending on the volume of the absorbent load. Therefore, to avoid excessive absorbent losses, it is essential to obtain extremely high absorbent regeneration ratios for each cycle (>99%), which is a genuine challenge considering the large number of impurities contained in coal exhaust gases, including SO_2 and NOx, oxygen due to parasite air entries and excess air for combustion. At the present time, no purification operation has been demonstrated with an efficiency of more than 99%. The global efficiency of regeneration of the absorbent will depend firstly on the efficiency of thermal regeneration, and secondly on chemical regeneration (treatment of salts on resins).

4.1.6. Limitations on operating flexibility of power stations with capture

Obviously, the CO_2 capture operation increases the complexity of power stations and in particular creates a problem of management of the interface with the downstream side transport and storage systems. This aspect has not been studied in detail and it will have to be sooner or later before any demonstration is possible. It will then be necessary to setup fallback procedures for the transport system or the storage system, in case the power station is shutdown.

4.2. Economic limitations

The economic estimates presented in this balance are relevant for a comparison between the different systems, but their absolute values may vary significantly as a function of the cost of metals, the price of fuels, tension on the equipment market (availability of construction and supervision teams), etc. For example, prices of metals have increased continuously since 2002 - 2003, to double by 2007 - 2008, which obviously has a large impact on the final investment and production cost, but should not modify relative costs between different systems.

Similarly, if the discount rate changes, it will have a significant impact on all economic estimates: the investment cost, the production cost and the cost per tonne of CO_2 avoided. As we have seen, the fuel cost also has an important impact on the production cost and on the cost per tonne of CO_2 avoided, particularly for NGCC for which it represents more than 70% of the production cost. In some cases, our estimates are very similar to estimates made by our American and Canadian colleagues, although the structure (breakdown) is significantly different. Fuel, equipment and installation prices are usually lower in North America, but discount rates may be higher than in Europe [12-14].

Concerning IGCC, it is clear in our studies that dry pressurization of coal is one of the key elements that can significantly improve the technical and economic performances of this technology. Progress on dry coal pressurization pumps is necessary, to avoid firstly the complex lock systems in the case of a Puertollano type IGCC, and the mixture of coal with water in the case of the IGCC-slurry; the recent announcement about development of such a pump capable of achieving a pressure of 40 bars suggests a potential improvement for this type of technology.

Finally, the question about operation at low loads, at semi-base or at peak-load should be considered. In theory, all systems could operate at a minimum load of 50% and for a number of hours corresponding to operation in semi-base or in peak-load. However, efficiencies at low load are seriously degraded and production costs, that are already high for CO_2 capture operations, increase significantly if the power station does not operate at base load. Thus, for an annual operation of only 3750 hours instead of 7500 hours, the cost of an IGCC-slurry with capture increases by almost 60%, which is more than double the cost of the IGCC-slurry without CO_2 capture. The same calculation for PC gives a production cost with oxy-combustion capture increased by 63% for 3900 hours of operation per year, compared with the cost of production for 7800 hours of operation per year. The impact on the production cost of operation in semi-base for NGCC is lower than for IGCC and PC because the proportion of the investment in the production cost is lower, and the cost increases by 24% in NGCC with post-combustion capture when operating for 4000 hours per year instead of 8000 hours per year.

5. Conclusions

The technical and economic estimates presented in this work are not sufficient alone to determine the choice of a system for the capture of CO_2. Constraints related to resources such as fuel and water may also influence this choice. Therefore, it appears difficult to make a once-and-for-all decision about the “best CO_2 capture process” ; however the results of our studies suggest that pre-combustion capture by physical absorption (methanol) should be used for IGCC, oxy-combustion should be used for PC and post-combustion capture (amines) should be used for NGCC.

However, these recommendations are based on a number of assumptions that may be more or less "dependable" depending on the process. For example, capture processes are supposed to operate reliably, which remains to be demonstrated on an industrial scale, at least for post-combustion and oxy-combustion capture. On the other hand, pre-combustion capture on synthetic gas is industrially mature, but the IGCC system into which it must be integrated still has availability problems. The studies that we have carried out assume that the specific problems that we will encounter for coal-fired power stations with capture have been solved, which is not the case at the present time: coal combustion boiler with oxygen, hydrogen turbine for IGCC with CO_2 capture, degradation of amines, separation of incondensable gases in oxy-combustion (and what about impurities in a cryogenic system?), etc. Finally, it is quite clear that the evaluation studies that we have carried out are no more than a first phase and must be continued with other more detailed pre-feasibility studies, particularly in engineering, to enable a more accurate evaluation of the industrial feasibility of the different processes for capturing CO_2.

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